Clean Air Research Program
Air Quality Summary and Abstracts

May 2009

Session 2: Air Quality

1.0 Introduction

The research described in this overview supports Long Term Goal 1 of the Clean Air Research Multiyear Plan by providing methods, measurements, and models to support the implementation of air pollution regulations. These methods, measurements, and models provide key insights into source emissions, atmospheric processes, and ambient concentrations and the relationships among these elements of the source to outcome paradigm. Thus, the research also supports Long Term Goal 2 by providing guiding measurements and insights across the source to outcome paradigm. As outlined in the Clean Air Research Multiyear Plan (MYP), the key science questions being addressed by Air Quality research are:

- What are the characteristics of air pollutant emissions from different types of sources and how do transformations in the atmosphere affect air pollutant concentrations and human exposures?

- What are the expected future concentrations of air pollutants and how can we evaluate and manage their potential adverse consequences?

The research described in this overview has lead to the completion of Annual Performance Goals (APG) in the MYP and will contribute to the completion of future APGs (2010 and beyond). The products of this research are used by air quality managers at the federal, state, and local levels to develop and evaluate emission reduction strategies designed to meet national air quality regulations and to measure compliance with these regulations. In addition, the tools supported by this research are being extended to provide important insights into ecosystem impacts, global climate change, and human exposure assessment.

2.0 Research Drivers

Air Quality research is driven primarily by the requirements and challenges associated with implementation of NAAQS. As noted in the Multiyear Plan, some specific examples of these challenges include:

- Continuing nonattainment problems (particularly for PM and Ozone),
- Uncertainties around predicting impacts of control strategies on air quality,
- Development of improved methods to effectively and rapidly measure pollutants,
- Uncertainties around the input data and atmospheric processes for refinement of air quality models,
- Uncertainties around which sources contribute to ambient levels of PM; and ,
- Development of improved emission inventories.

In addition, two NRC reports published in 2004 also provide important insights for directing air quality research. The fourth NRC Report on Research Priorities for Airborne Particulate Matter identified the following challenges related to air quality research:

- Completing the PM emissions inventory and PM air quality models necessary for NAAQS implementation and for informing health research.
- Enhancing air quality monitoring for research
- Integrating disciplines
- Moving beyond PM to a multi-pollutant approach
- Accountability

The NRC Report on Air Quality Management in the United States included the followed recommendations related specifically to air quality research:

- *Improve emissions tracking*, including new emissions monitoring techniques and regularly updated and field-evaluated inventories.
- *Enhance air pollution monitoring*, including new monitoring methods, expanded geographic coverage, improved trend analysis, and enhanced data accessibility.
- *Improve modeling*, including enhanced emission and air measurement programs to provide data for model inputs and model evaluation and continued development of shared modeling resources.

The Air Quality research in ORD's Clean Air Research Program is making progress in addressing the challenges and recommendations above. However, to address several of these challenges, in addition to others identified in the NRC reports, will require continued integration and coordination among all the research elements in the Clean Air Research Program.

3.0 Research Summary

The following sections provide a brief summary of the Clean Air Research Program's Air Quality research efforts related to source emissions, atmospheric measurements, and air quality modeling. This research provides important insights about the relationships between source emissions and ambient concentrations and the potential for exposures to humans and ecosystems.

3.1 Source Emissions: Estimation and Evaluation

ORD conducts source emissions research to improve the understanding of sources contributing to air quality problems with a primary emphasis on improving emission inputs needed for air quality models, such as the Community Multiscale Air Quality (CMAQ) model. This research uses various sampling and analytical techniques to collect and analyze measurement data to provide important insights into the magnitude, composition, and atmospheric fate of source emissions. Much of the recent source emission research has been focused on widely dispersed or uncontrolled sources, such as wild/prescribed fires, biogenic sources, and agricultural/natural sources of ammonia. Due to the dispersed nature of these sources, innovative measurement techniques are being deployed, such as satellite imagery and passive methods.

ORD is working with the U.S. Forest Service and NASA to develop and employ more robust fire emission algorithms and incorporating satellite imagery to improve the temporal and spatial resolution of biomass burning emissions. In the area of biogenic emissions, ORD researchers have also improved the experimental protocols and measured the emissions of isoprene, sesquiterpenes, and monoterpenes from a variety of tree species. They have also determined the

temperature and light dependence of these emissions, which is essential to understanding how the emissions contribute to particulate matter concentrations under changing climatic conditions. Isoprene, which is the atmosphere's second most abundant hydrocarbon and emitted almost entirely by vegetation, was identified by ORD researchers as a significant source of organic aerosol particles. Finally, ORD's ammonia emissions research has provided key insights into bi-directional flux in both agricultural and natural settings and demonstrated that only a small fraction of ammonia emissions from animal production facilities deposits locally.

ORD is also conducting research to improve characterization of anthropogenic sources of aerosols through improved sample collection and analysis techniques. ORD's source emissions research program also includes a mobile source component, which has contributed data on emissions distribution and speciation from on-road light duty vehicles, emissions from laboratory tested vehicles using ethanol blended fuels, and emissions from small non-road engines, such as lawn and garden equipment. Finally, ORD is also working to integrate measurements and modeling tools using inverse modeling techniques to evaluate and improve emissions estimates.

Table 1 identifies the Annual Performance Goals (APG) in LTG 1 of the MYP to which the Source Emissions research described above contributes.

Table 1. Source Emissions Research Contributions to the Clean Air Research MYP

APG #	APG Description	Date	Contributions through 2009
APG 9	Provide advanced air quality models that incorporate the latest atmospheric and emissions data to OAR and States	2008	Characterization of emissions from wild and prescribed fires
APG 10	Deliver new and improved techniques to measure and characterize source and ambient concentrations of PM and PM-related precursors and toxics	2009	Ammonia measurements
APG 11	Provide models, data, and tools to better manage PM in the atmosphere, including carbonaceous particles that contribute to high levels of PM in areas of the country where NAAQS attainment will linger even after national rules are implemented.	2011	Novel methods to characterize combustion emissions
			Integration of receptor, source-based, and inverse models for PM source apportionment
APG 13	Provide new emissions factors and chemical composition data for dispersed sources of air pollutants, including off-road vehicles (airplanes, ships, construction equipment) to directly support State efforts to improve emissions inventories	2011	Chemical characterization as a function of particle size
			Enhance and update SPECIATE database

3.2 Ambient Measurements: Air Quality Characterization and Process Insights

Ambient measurements support numerous air quality management activities, such as determining compliance with NAAQS, characterizing ambient concentrations, identifying relative source contributions, and providing atmospheric process insights to improve air quality modeling tools. ORD's ambient measurements research program includes the development and evaluation of sampling and analytical methods and the application of measurement methods to provide insights into source contributions, atmospheric processes, and ambient concentrations.

The Clean Air Research Program's methods development research includes the development and evaluation of Federal Reference and Equivalency Methods (FRM/FEM) needed to measure compliance with NAAQS, as well as, research to develop and evaluate advanced methods needed to enhance other air quality management activities. ORD coordinates with the Office of Air and Radiation (OAR) in the development and evaluation of FRMs required for NAAQS monitoring and compliance. To enhance this coordination, ORD has recently identified ambient method contacts for each criteria pollutant who serve on OAQPS teams supporting the review process for each NAAQS. In addition, ORD develops testing and acceptance criteria and reviews applications for FEMs, which may also be used to determine compliance with NAAQS. A recent example is the influx of FEM applications for continuous fine particle mass methods in response to new testing and acceptance criteria included in the 2006 PM NAAQS. As approved continuous particle mass FEMs are deployed in the national monitoring network they may provide valuable new insights for development and implementation of NAAQS, while continuing to provide compliance data.

In addition to the development and evaluation of FRM/FEMs, ORD is also developing, and evaluating other ambient sampling and analytical methods that support air quality management activities such as source apportionment and air quality modeling. The ambient measurement methods being developed and evaluated by ORD include continuous methods for PM species, passive methods (for coarse particles, ammonia, and air toxics), mercury measurement methods, and portable optical techniques. ORD's research program also includes the development and evaluation of advanced analytical methods, including methods to identify organic tracer compounds and elemental carbon.

The Clean Air Research Program's ambient measurements research also includes the application of methods to provide important insights. For example, the integrated application of high time resolution sampling methods, sensitive analytical methods, and receptor modeling tools provides new opportunities to improve the understanding of relative sources contributions. This approach has been applied in several urban areas to identify sources of PM and mercury. In addition, measurement and modeling studies have provided new insights into mercury fate and transport, such as the role of high altitude oxidation and mechanisms of bi-directional flux. Ambient and laboratory measurements have also contributed to an enhanced understanding of the precursor species and atmospheric mechanisms that lead to the formation of secondary organic aerosol (SOA). Results from ORD work have shown how particles form from cycloalkenes, organics, and other volatile organic compound precursors. The previously unrecognized importance of particle acidity, the presence of nitrogen oxides, and the formation of oligomers have been demonstrated. Incorporation of these advances in understanding will improve how well regional air quality models represent particle concentrations in the atmosphere and increase the ability of planners to use these models to predict air quality and develop plans for reducing particulate matter concentrations. Finally, atmospheric measurements are also providing new insights into the spatial variability of coarse particle ambient concentrations within urban areas, as well as, the regional compositional variability of coarse particles.

Table 2 identifies the Annual Performance Goals (APG) in LTG 1 of the MYP to which the Atmospheric Measurements research described above contributes.

Table 2. Atmospheric Measurement Research Contributions to the Clean Air Research MYP			
APG #	**APG Description**	**Date**	**Contribution**
APG 9	Provide advanced air quality models that incorporate the latest atmospheric and emissions data to OAR and States	2008	Improved SOA chemistry module
APG 10	Deliver new and improved techniques to measure and characterize source and ambient concentrations of PM and PM-related precursors and toxics	2009	Verification of portable optical devices
			Passive methods for ammonia and coarse particles
			Analytical methods for organic speciation, EC/OC, and tracer compounds
APG 11	Provide models, data, and tools to better manage PM in the atmosphere, including carbonaceous particles that contribute to high levels of PM in areas of the country where NAAQS attainment will linger even after national rules are implemented.	2011	Improved SOA formation mechanisms
			Improved linkages between emission sources and atmospheric processes for primary carbonaceous PM and SOA

3.3 Air Quality Modeling: Applications Driving Development and Evaluation

Air quality models are used by air quality managers at the national, state, and local levels to develop and evaluate policies and emission reduction strategies designed to protect humans and ecosystems from adverse impacts associated with exposure to air pollutants from various sources. ORD's air quality modeling research program develops, evaluates, and applies modeling tools with the overall goal of enhancing these tools to improve their reliability and credibility.

Much of the effort since 2005 to enhance air quality models has focused on particulate matter, with significant emphasis on the formation of secondary organic aerosols (SOA). Laboratory and field studies have yielded new insights regarding mechanisms for SOA formation, and air quality modeling researchers have conducted studies to incorporate and test these findings which resulted in better predictions of SOA. Within the CMAQ model, the role of clouds in SOA formation has been incorporated using new mixed-phase chemical mechanisms that resulted in further improvements in SOA predictions. New insights regarding the semivolatile nature of primary organic aerosol (POA) has lead to the development of the Volatility Basis Set (VBS) approach that permits models to efficiently treat both semi-volatile POA and SOA and improves model simulations of organic aerosol. This work transforms scientific thinking and representation of organic aerosols and suggests that significant changes are needed in current approaches to more accurately measure and regulate emissions contributing to particulate matter in the atmosphere. Preliminary versions of the VBS approach have been implemented in both the CMAQ and CAMx modeling systems and resulted in improved performance. Other PM modeling research has lead to enhanced treatment of coarse and ultrafine size fractions. For coarse particles, modeling enhancements were made with respect to the treatment of inorganic gases, crustal material, and sea salt. Modeling ultrafine particles has been improved through capturing regional nucleation events and estimating the size distribution of emitted combustion particles.

While improving model performance for particles has been a major emphasis since 2005, other modeling advancements have also been made. Multipollutant modeling has improved such that the CMAQ model can now simultaneously predict concentrations of ozone, particulate matter, nitrogen oxides, lead, and 43 hazardous air pollutants (HAPs), including mercury. Multimedia modeling has also improved as a result of including interactions between gas- aqueous- and particle-phase chemistries and linking the CMAQ model with watershed models. In addition, the meteorological inputs to air quality models have been improved with the transition to the state-of-the-science Weather Research and Forecast (WRF) model from the older mesoscale modeling system MM5. The transition has included incorporation of improved land-surface processes and planetary boundary layer algorithms into WRF. Finally, the reliability and credibility of air quality models has been tested and improved through a comprehensive model evaluation framework that includes operational, diagnostic, dynamic, and probabilistic approaches.

In addition to the source-based air quality modeling advances, receptor-based air quality modeling tools have also been developed and evaluated in the Clean Air Research Program. EPA versions of the Unmix and Positive Matrix Factorization (PMF) models have been developed and made available via the internet. User guides have been developed and training sessions have been conducted to educate potential users, including state and local agencies, about the use of receptor-based modeling tools. These tools have also been applied in several urban areas (e.g., St. Louis, MO, Dearborn, MI, and Steubenville, OH) to identify sources contributing to air pollution problems.

Table 3 identifies the Annual Performance Goals (APG) in LTG 1 of the MYP to which the Air Quality Modeling research described above contributes.

Table 3. Air Quality Modeling Research Contributions to the Clean Air Research MYP

APG #	APG Description	Date	Contribution
APG 9	Provide advanced air quality models that incorporate the latest atmospheric and emissions data to OAR and States	2008	CMAQ release with improve SOA mechanisms
			Release of EPA Versions of Receptor Modeling Tools (PMF and Unmix)
APG 12	Develop modeling systems that couples air quality and meteorology models for better estimates and forecasts of ambient ozone and PM2.5	2011	Public release of coupled interactive WRF-CMAQ modeling system
			Analysis and evaluation of PM forecasts using CMAQ
			Public release of next version of CMAQ model for addressing residual non-attainment problems for ozone and PM2.5

3.4 Extending Applications of Air Quality Management Methods and Models

Historically, Air Quality research has focused on delivering methods, measurements, and models that support implementation of primary NAAQS, particularly PM and ozone. However, the application of these methods, measurements, and models is now being extended to other areas of air quality management. First, the CMAQ model has been used to assess impacts of NOx and SOx on ecosystems and is directly supporting the
current multi-pollutant review of the secondary NAAQS for NOx and SOx. Ecosystem modeling efforts have identified the importance of regional sources of nitrogen and sulfur and the important role of ammonia in ecosystem deposition. Second, emissions and air quality

modeling tools are being used to evaluate potential impacts of climate change. Results from air quality modeling runs have shown significant impacts on increased ozone concentrations and a lengthening of the ozone season resulting from projected climate change scenarios. New emission scenarios are being developed that more comprehensively account for projected changes in a variety of factors, such as population, economics, and land use. Finally, air quality modeling tools are being developed and applied to improve our understanding of human exposures to air pollution. These modeling approaches are providing more spatially refined estimates of air pollution, which may be used alone or as an input into human exposure models, to provide estimates of exposure for use in health studies. The research to extend air quality methods and models will contribute to future APGs in both LTGs 1 and 2 in the Clean Air Research MYP.

4.0 Impacts and Outcomes

The Air Quality research summarized above has resulted in significant positive impacts and outcomes. First, the research has advanced air quality science by providing new insights regarding emissions from a variety of sources (e.g., biogenics, fires, ammonia, and primary organic aerosols) and atmospheric processes (e.g., SOA). These scientific advances have lead to improvements in tools used by air quality managers, including the National Emissions Inventory (NEI), the Multiscale Motor Vehicle and Equipment Emission System (MOVES), and the CMAQ model. The enhanced CMAQ model, in particular, has been used directly to inform development of air quality management policies including:

- Clean Air Mercury Rule (CAMR)
- Clean Air Interstate Rule (CAIR)
- Renewable Fuel Standards (RFS)

This research has also directly supported implementation of NAAQS by providing methods to determine compliance, as well as, tools and data for developing State Implementation Plans (SIP) for non-attainment areas. Table 1 provides a summary of the FRM and FEM designations since 2005.

Designation	Number of Designations by Pollutant Since 2005							
	PM_{10}	$PM_{2.5}$	$PM_{10-2.5}$	O_3	NO_2	SO_2	CO	Totals
Federal Reference Methods (FRMs)	1	3	3		3		3	13
Federal Equivalent Methods (FEMs)	2	10	2	4		3		21
Modifications to FRMs	9	3		2	5	8	8	35

Table 4. FRM and FEM Designations Since 2005

All of the designated FRM and FEM are now available to measure compliance with NAAQS in the national monitoring network, including the use of new FEMs for continuous measurement of fine particles. In addition to compliance methods, ORD's research has supported development of SIPs by providing tools and data. The CMAQ model and receptor modeling tools, such as PMF and Unmix, have been made available to and are being used by state and local air quality managers. Figures 1 and 2 provide data on users of these modeling tools.

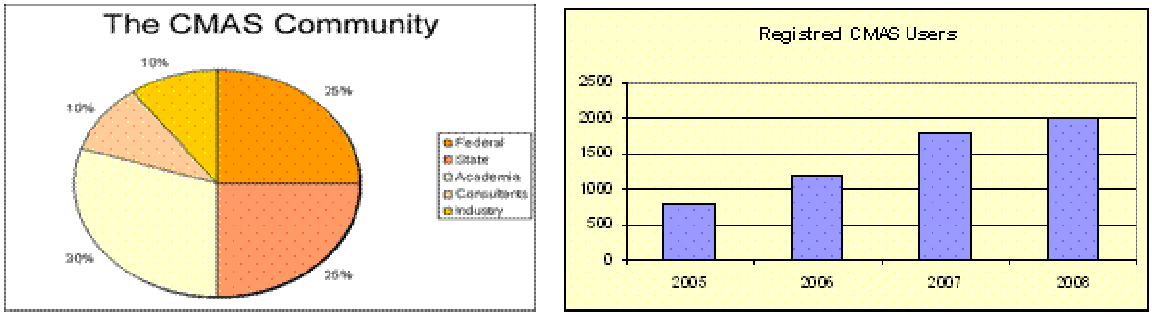

Figure 1. Community Modeling and Analysis System (CMAS) Users (US and International)

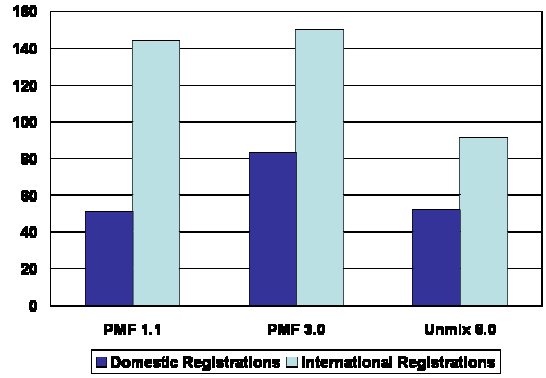

Figure 2. PMF and Unmix Registrations in the US since September 2007

Figure 1 provides summary statistics from the Community Modeling and Analysis System (CMAS) center. The CMAS center, a collaborative effort with the Institute for the Environment at University of North Carolina-Chapel Hill, facilitates and promotes the use of several modeling tools including CMAQ. As shown in Figure 1, there are approximately 2,000 registered CMAS users in the US and internationally that come from a variety of backgrounds, including federal, state, academia, and industry. Figure 2 provides information on the number of registered users in the US for the PMF and Unmix models since September 2007. The total number of US registrations is 186 and there are an additional 385 international registrations. While these numbers are lower than the CMAS registrations, it is important to note that these models have been available for download from the EPA website for less than two years. In addition to modeling tools for SIP development, ORD studies in St. Louis, MO and Dearborn, MI have provided data on relative source contributions in non-attainment areas and a similar study is planned for Cleveland, OH.

Finally, ecosystem modeling analyses have directly supported air quality management activities related to the Cheasapeake Bay restoration efforts and Tampa Bay Estuary Program. ORD's development of the Watershed Deposition Tool has greatly facilitated the use of air quality modeling data by watershed managers.

5.0 Future Directions

The Air Quality research in the ORD's Clean Air Research Program will continue to support implementation of air regulations through the development of methods, measurements, and models to characterize source emissions, atmospheric processes, and ambient concentrations and to understand relationships between source emissions and ambient concentrations. Future

contributions to LTG 1 of the Clean Air Research MYP include enhanced emission characterization for a variety of sources (e.g., aircraft engines, off-road mobile sources, and mobile sources using alternative fuels), new releases of the CMAQ model, a coupled interactive WRF-CMAQ modeling system, fine scale and global scale air quality modeling tools, and enhanced receptor-based modeling tools.

Consistent with the strategic transition in LTG 2 of the MYP, the tools and products produced from Air Quality research will continue to be extended to support multi-pollutant research, source-to-outcome objectives, and accountability. Many of the air quality tools already have multi-pollutant capabilities. For example, the CMAQ model was used by OAQPS in the Detroit Multi-pollutant Pilot Project. These multi-pollutant tools will continue to evolve to support additional multi-pollutant analyses. In addition, the renewed emphasis on the 5-year NAAQS review cycle for all criteria pollutants also calls for expanding the multi-pollutant capabilities of Air Quality research since analyses supporting individual NAAQS will need to consider multi-pollutant impacts. Source-to-outcome will also be supported by Air Quality research. The enhanced characterization of ambient concentrations through the use of air quality models will lead to improved exposure assessments for health studies, as will the source apportionment techniques. Finally, as called for in the NRC Report on Air Quality Management in the US, the concept of air quality management should be revised to an iterative process of standards setting, standards implementation, and assessment of progress. This concept of "accountability" is presented in figure 3 from the NRC Report. Air Quality research in ORD will provide and enhance the science and technical foundation for this process through source emissions, atmospheric measurement, and air quality modeling research and the evaluation of methods for assessing progress toward public health and ecosystem protection objectives.

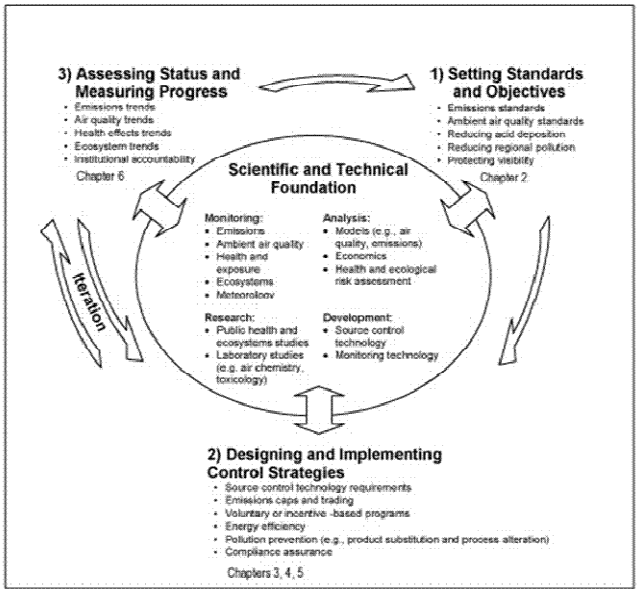

Figure 3. Iterative Nature of Air Quality Management
(Source: NRC "Air Quality Management in the US, "2004)

Poster # LTG 1-20

How is Our Evolving Understanding of Biogenic Emissions Helping to Represent Their Role in Multipollutant Atmospheric Chemistry?

Presenter: Christopher D. Geron, US EPA Office of Research and Development

Poster Summary: It has been widely shown that biogenic volatile organic compound (BVOC) emissions are important in tropospheric ozone chemistry, and recently quantitative estimates of BVOC contribution to secondary organic aerosol formation (SOA) in the eastern U.S. have been documented (Kleindienst et al. 2007, Geron 2009). Since the 1980s, on roughly a five-year cycle, ORD collaboration with the National Center for Atmospheric Research (NCAR) and Washington State University has yielded a progression of steadily improving models of reactive trace gas emissions from biogenic sources. The most recent is MEGAN (Model of Emissions and Aerosols from Nature, Guenther et al. 2006). MEGAN is driven by biological, physical, and chemical variables derived from ground- and satellite- based observations as well as models. While studies have documented good performance of MEGAN globally and in many key forested regions, model comparisons with ambient data have identified several areas where additional data are needed. These include better temporal parameterization of temporal/phenological effects, improved emission factors and algorithms for compounds other than isoprene from non-forest ecosystems such as agricultural crops, grasslands, bioenergy fuel crops, and shrublands. In addition, several studies have provided indirect evidence that "missing" BVOC (unknown compounds that are currently not included in BVOC emission models) could contribute between 50 and 90% of the total BVOC emission. ORD and NCAR are addressing these needs by conducting trace gas and aerosol measurements at leaf to landscape scales using a variety of ambient and enclosure methods.

Impacts and Outcomes: MEGAN represents a significant advancement in natural emission models. Evaluations of MEGAN using ambient measurements often indicate good performance for isoprene emissions. In cases where significant discrepancies have been found, there is some evidence that the differences are due to a limited understanding of isoprene oxidation chemistry, especially in regions of low NO_X. Development and application of advanced measurement techniques have resulted in emission factor data included in MEGAN and flux estimates used to test the model at canopy to landscape scales. These include eddy covariance and relaxed eddy accumulation (REA) flux methods, PTR/MS systems on aircraft, and advanced techniques to manage analyte losses via oxidant reactions (e.g. Arnts 2008). New portable GC/MS, chemical ionization mass spectroscopy (CIMS), and chemical adsorbent technologies have been applied in emissions field studies of BVOC compounds (Geron et al.2006, Matsunaga et al. 2008, Arnts in prep.). These studies have resulted in new emission factor estimates for oxygenated BVOC from forests, sesquiterpenes from forests and agriculture, and other compounds such as UV-absorbing compounds from desert vegetation which have high SOA forming potential. Under the STAR program important findings have been published by Monson (2007), Kroll et al. (2006), and Lee et al. (2006). Among the findings are that leaf isoprene emission has been found to decline under increasing CO_2, which has important air quality implications under future atmospheric conditions.

Future Directions: Over the next five years work will focus on integration of MEGAN into CMAQ and additional model performance validation. ORD is collaborating with NCAR in developing landscape characterization data which will enhance the spatial, temporal, and chemical resolution of natural emissions for input into ORD air quality models. We will continue enclosure, canopy flux and laboratory emission factor studies of the biological basis for seasonal variation in BVOC emissions, including an emphasis on ecosystems other than forests. Currently, seasonal variability in emission estimates of important biogenic compounds is driven entirely by temperature response algorithms. Recent studies show that phenological events, extreme weather (e.g., storms, drought), and insect outbreaks can result in order of magnitude changes in BVOC emissions and therefore large errors in many current models. VOC –SOA interaction is still a developing research area and additional data on emissions of reactive and high aerosol yielding compounds (and their controlling factors) is needed. Emissions of compounds such as

methyl salicylate, methyl chavicol, and several sesquiterpenes have high aerosol yields and are correlated with stress or phenological events which can be simulated in future versions of MEGAN.

Relevant Publications

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Wiedinmyer, C., J. Greenberg, A. Guenther, B. Hopkins, K. Baker, C. Geron, P. Palmer, B. Long, J. Turner, G.Petron, P. Harley, T. Pierce et al. (2005) "Ozark Isoprene Study (OZIE): Measurements and modeling of the "isoprene volcano", Journal of Geophysical Research **110**(D18307):1-17.

Poster # LTG 1-21

What is the Significance of Emissions from Wildland and Prescribed Fires?

Presenter: Thomas E. Pierce, US EPA Office of Research and Development

Poster Summary: While wildland and prescribed fires are vital for ensuring the health of natural ecosystems and for maintaining the productivity of timber and agricultural lands, it is important to understand how biomass burning contributes to harmful levels of air pollution. Over the past five years, ORD has established a research portfolio to reduce the uncertainty in characterizing the emissions, transport, and transformation of gases and particles from wildland and prescribed fires. By examining speciated fine particulate ($PM_{2.5}$) concentrations over 3-years in central North Carolina, Geron (2009) concluded that biomass burning contributed as much as 40% to organic carbon (OC) and was responsible for extreme daily pollution events. To further characterize source profiles from biomass burning, Gullett et al. (2008) and colleagues have quantified the chemical composition for dioxins and other toxic chemicals such as benzene, toluene, naphthalene, and styrene for a variety of fuels and fire conditions. Hays et al. (2007) discerned the nanostructures of individual soot particles using high-resolution electron microscopy to help uncover the mechanisms underlying particle interception and growth and understand how soot nanoheterogeneity may contribute to $PM_{2.5}$-related health effects.

Satellite imagery has played a huge role in improving the understanding of fire emissions. Mazonni et al. (2007) report on use of the MISR instrument to build a database on the vertical distribution of major smoke plumes. Several groups (e.g., Widenmyer et al., 2006; Al-Saadi et al., 2008; Pouliot et al., 2008) have relied upon satellites to construct biomass burning emission inventories. Now, satellite-generated fire emissions are used routinely in air quality forecasts by NOAA and for retrospective analyses by EPA and its partners for air quality management decision-making. Soja et al. (2009) report that a satellite-based approach can capture ~90% of the total area burned by wildland and agricultural fires in North America. Work on satellite imagery to build emission inventories has benefited from partnerships with NASA and the U.S. Forest Service. Finally, burn-scar images from satellites like SPOT and LANDSAT have been used by researchers to "validate" estimates of burn areas. Despite the success with satellites, gaps still exist when temporal coverage is lacking and where fires burn in the understory (e.g., in the southeastern U.S.). Wang et al. (2007) offer encouraging results for detecting "cooler" fires with an improved algorithm using four thermal infrared (TIR) channels on the MODIS platform.

Research under the Global Climate program is yielding more sophisticated fire behavior models, especially those that may predict fire occurrence in a future climate state. Spracklen et al. (2007) analyzed OC concentrations from 1989-2004, and using a global chemical transport model (GEOS-chem), found that modeled OC concentrations were much more highly correlated with observations if fire-specific emissions (rather than climatological fire averages) are assumed ($r^2 = 0.8$ v 0.4). In concluding that increased wildfire activity in the western U.S. since the mid 1970s has resulted in a 30% increase in OC concentrations, they suggest that a warmer/drier climate could lead to increased fire activity and adversely impact $PM_{2.5}$ concentrations and visibility.

Impacts and Outcomes: Research results in this program area are leading to an improved understanding of how biomass burning emissions contribute to atmospheric pollutant levels. Use of satellite imagery has resulted in major improvements to EPA's National Emissions Inventory (NEI), which is a cornerstone for air quality decision making. Estimated fire emissions in the NEI have evolved from a crude, state-based, monthly average to a fire-specific, daily-resolved estimate. EPA's external research partners are providing improved knowledge in areas such as chemical composition of smoke, plume rise measurements via satellites, and the development of improved emission algorithms. This information is being used to support the development and implementation of National Ambient Air Quality Standards, especially for PM and ozone.

Future Directions: Over the next few years, ORD will be integrating advances made in the external research programs into EPA's emission modeling systems, particularly in the areas of chemical speciation, emission factors, plume rise, fuel loading, and fire behavior modeling. ORD plans to work with the EPA program offices and other Federal land managers to ensure a coordinated integration of these tools. Finally, ORD plans to assess air quality model performance against field observations using these improved tools and databases.

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Poster # LTG 1-22

How Do We Quantify Emissions of Ammonia From Agricultural and Natural Sources?

Presenter: John Walker, US EPA Office of Research and Development

Poster Summary: Animal manure and fertilized soil are the primary sources of atmospheric ammonia (NH_3). While previous work in ORD focused on quantifying NH_3 emissions from animal production, recent work focuses on reducing uncertainty in emissions from fertilized and natural soils. Because overlying foliage may act to reduce or increase net emissions to the atmosphere, this work also investigates the rates and processes of NH_3 emission and uptake (i.e., deposition) by vegetation. These aspects of NH_3 biogeochemical cycling remain poorly characterized, primarily due to a paucity of measurements of sufficient quality and process-level detail. Such data are needed to develop more accurate and mechanistically representative models of NH_3 emission from fertilized soils and bi-directional air-surface exchange with natural soils and vegetation. Because NH_3 is a precursor of inorganic aerosol, uncertainty in air quality model predictions of net NH_3 air-surface fluxes in agricultural and natural landscapes translates to uncertainty in predicted atmospheric particulate matter (PM) concentrations. Furthermore, this uncertainty also prohibits accurate quantification of total nitrogen deposition budgets for sensitive ecosystems and, therefore, the development of an effective "critical load" mitigation approach. Further development and evaluation of PM control and critical loads strategies to protect human and ecosystem health requires more accurate air quality model (AQM) predictions of NH_3 air-surface exchange with agricultural and natural soils and vegetation. ORD is addressing this issue by using measurements and modeling to quantify component (i.e., soil and foliage) and net air-surface fluxes of NH_3 from crops and natural ecosystems at field to regional scales.

Impacts and Outcome: Field studies were conducted in 2006, 2007, and 2008 to characterize NH_3 fluxes and surface processes in fertilized corn, soybean, and unfertilized pasture. Spectroscopic and online wet chemical techniques were employed to quantify air-surface exchange using micrometeorological approaches. Passive sampling techniques were used in a routine monitoring framework to characterize the spatial variability of atmospheric NH_3 concentrations in eastern North Carolina where animal agriculture is widespread. Measurements show that net canopy-scale fluxes are bi-directional and the rate of emission or uptake depends on surface wetness, foliage nitrogen status, and atmospheric NH_3 concentration. The majority of NH_3 emitted from fertilized soil is recaptured by the overlying canopy. Air concentration measurements and inferential modeling demonstrate that 5 - 15% of the NH_3 emitted from animal production facilities is recaptured by vegetation within 500 m; thus the majority of emissions are available for PM formation and deposition to more distant receptors. This finding is consistent with CMAQ model budget simulations, which show that 15 – 30% of hot spot emissions are deposited within the same 12x12 km grid cell. Field measurements have been used to develop improved algorithms for simulating in-canopy air flow, NH_3 emissions from fertilized soils, and bi-directional canopy-scale fluxes over crops. Incorporation of these algorithms into CMAQ will improve the spatio-temporal resolution and accuracy of estimated agricultural NH_3 fluxes and their impact on regional air quality.

Future Directions: Future efforts to address this question will focus on transferring new soil NH_3 emission and bi-directional canopy-scale flux algorithms to the CMAQ air quality model. Additional field studies will be conducted to quantify NH_3 fluxes in forest canopies and further characterize the spatial and temporal variability of atmospheric NH_3 concentrations in agricultural areas. Data from these studies will be used to evaluate CMAQ predictions of NH_3 emission and fate and to validate NH_3 retrievals from the TES-Aura satellite.

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Poster # LTG 1-23

How Are Source Sampling and Characterization Techniques Evolving to Measure Criteria and Toxic Air Pollutants Emitted From Anthropogenic Combustion Sources?

Presenter: Michael D. Hays, US EPA Office of Research and Development

Poster Summary: There is considerable interest in developing and improving source emissions collection techniques and the analyses used to discern the dynamic and complex nature of anthropogenic aerosols. Several key advancements related to anthropogenic $PM_{2.5}$ sample collection and analysis were made recently. These include advances in (i) understanding the limits facing dilution sampling measurements with regard to primary organic aerosols (POA), (ii) off-line instrumentation for providing organic chemistry by particle size, (iii) metals analysis, (iv) natural sources and biomass burning emissions chemistry, and (v) continuous or real-time single particle instrumentation. In addition, atmospheric studies in near-source environments are now confirming emissions testing results and deriving more representative emissions factors. The research on anthropogenic emissions to be presented supports, tests, or changes key assumptions implicit in (i) PM standards implementation, (ii) air quality modeling and analysis, (iii) emissions inventory development, (iv) the interpretation of health studies, (v) understanding the effects of atmospheric constituents on visibility impairment, or (vi) air monitoring network design. The results being reviewed here were produced over a variety of research institutions including EPA-ORD, Carnegie Mellon University, Arizona State University, University of California-Davis and -San Diego, University of Southern California, and University of Wisconsin-Madison.

Impacts and Outcomes: (1) POA emission factors have been biased high owing to low to moderate dilution in widely utilized sample collection and measurement systems. Although historically treated as non-volatile, in the atmosphere POA emissions are largely gas-phase. Consequently, input to chemical transport models was revised to account for the formation of secondary organic aerosols (SOA) from anthropogenic emissions sources. (2) New techniques coupling impactor-based particle collection with off-line chromatography show that traditional organic markers may be utilized in the ultrafine particle size range and that much of the organic matter resides in particles with aerodynamic diameters of 1 micrometer or less. (3) Determination of toxic metal speciation and valence states in PM with novel fine structure and mass spectrometry methods is improving health effects knowledge. (4) In regions across the U.S., the substantial contribution of primary biological air particles to residual non-attainment areas is becoming clearer with the development of a program that studies the major saccharide compounds from bio- emissions. (5) Real-time single particle characterization is providing heretofore unavailable information about transient emissions events, the size and chemical composition of PM, and the effects of atmospheric dilution on source emissions. In the process, this instrumentation has improved our understanding of acute health effects associated with PM emissions events and the influence of fossil fuel combustion and source operating conditions on the size and composition of PM emissions. (6) Finally, the straightforward estimation of PM emissions factors from near-source environments will help overcome challenges related to dilution sampling, better reconcile the emissions data utilized for science-based policy and rule development, and determine the effectiveness of emissions controls.

Future Directions: Future research objectives include (1) developing source testing methods that improve the identification of SOA formed from vapor phase primary emissions; (2) introducing more sophisticated hyphenated chromatography techniques that advance our understanding of organic matter and biological source contributions to size-segregated atmospheric aerosols; (3) further examining the underlying role of metals in aerosol-mediated reaction chemistry and studying the various organically bound metal species; (5) deploying semi-continuous and real-time instrumentation to measure ultrafine emissions from highly efficient fossil fuel combustion sources not amenable to filter-based measurements and artificial dilution; and (6) continuing to reconcile National emissions data with near-source ambient measurements.

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Poster # LTG 1-24

How Can Measurements and Modeling Tools Be Used to Improve Emissions Estimates?

Presenter: Ted Russell, Georgia Institute of Technology, School of Civil and Environmental Engineering
Sergey Napelenok and Robert Pinder, US EPA Office of Research and Development

Poster Summary: Central to effective air quality management is the ability to accurately estimate emissions and quantify their impact on air quality. Emissions are seldom actually measured except for major facilities (e.g., power plant emissions of SO_2 and NO_x). Typically, they are estimated using a "bottom up" approach, starting with estimates of source activity, multiplied by estimates of the emissions per unit activity. Such an approach is subject to many uncertainties and potential biases.

In recent work, both the models and measurements have advanced substantially. The types of models used generally fall in two categories: receptor-oriented (e.g., the Chemical Mass Balance (CMB) or Positive Matrix Factorization (PMF)) and emissions-based (e.g., CMAQ), each having their strengths and weaknesses. Recent work has led towards combining the approaches using inverse modeling where results from emissions-based models identify how emissions estimates should be altered to provide results consistent with measurements. This approach has been applied to compounds impacting not only traditional air pollutants (e.g., CO, NO_x, PM, SO_2, NH_3 and VOCs), but also global pollutants as well (e.g., CO_2). This is a particularly exciting area as satellite observations of trace species are now available, providing widely-expanded spatial coverage, particularly in remote regions and developing countries.

Direct measurements are also important for better characterizing actual emissions because laboratory measurements are subject to operating conditions that may not represent those experienced in the field. For example, a forest fire typically burns very little wood, instead burning leaves, needles and duff, with incomplete combustion and VOC vaporization playing a larger role. Coordinated ambient measurements can provide better "real-world" characterization of source emissions. Measurements of particulate matter (PM) composition suggest that there is a seasonal change in the organic emissions from gasoline fueled vehicles and that prescribed burning emissions are much richer in VOC emissions, leading to unexpected increases in both ozone and secondary organic aerosol formation.

Impacts and Outcomes: Integrating ambient measurements and models have significantly improved emission estimates and continue to advance our ability to effectively manage air quality. Biases in ammonia and VOC emissions have been identified, and appropriate corrections are being implemented. Results for other compounds are less definitive.

Future Directions: Both the methods and measurements continue to evolve, suggesting that future estimates will improve further, and that the techniques can be applied to other sources and areas, e.g., in remote regions and developing countries. Inverse modeling approaches will be applied to the growing body of routine PM species observations to identify potential improvements in what is believed to be one of the most uncertain, but important, components of the current emissions inventories. Particularly exciting is the availability of satellite observations, but the tools and understanding for appropriately using such data must be developed as well.

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Poster # LTG 1-25

How Does Ambient Measurement Methods Research Support Development and Implementation of Air Quality Regulations?

Presenter: Robert Vanderpool, US EPA Office of Research and Development

Poster Summary: The adverse health and ecological effects associated with exposure to air pollution has long been recognized and led to the development of air quality regulations to restrict source emissions to attain acceptable levels of ambient air pollutants. Since the 1971 enactment of National Ambient Air Quality Standards (NAAQS) for the criteria pollutants, ORD has played a critical role in developing and evaluating both Federal Reference Methods (FRMs) and Federal Equivalent Methods (FEMs) for each of the NAAQS pollutants. While developed primarily to support NAAQS attainment determinations, these methods are also designed to support multiple monitoring objectives such as trends analysis and the ability to issue timely public health advisories. A recent example of ORD's contributions to air quality regulatory support was the successful 2006 promulgation of a new FRM for the coarse fraction of PM_{10} (i.e., $PM_{10-2.5}$) as well as development of testing requirements and acceptance criteria to enable introduction of continuous $PM_{2.5}$ and $PM_{10-2.5}$ FEM monitors into routine operating networks. Many of these specifications were developed based on ORD's comparison testing of both integrated and continuous PM monitors at multiple field sites. ORD was also responsible for the 2008 revisions to the Pb NAAQS which involved advances in both PM sampling technologies and Pb-specific analytical techniques. Overall, these regulatory revisions have enabled instrument manufacturers to make available to monitoring agencies a wide variety of improved measurement technologies. Since 2004, ORD has formally approved 17 new pollutant monitors for NAAQS compliance determinations, and an additional 15 new monitor applications are currently undergoing ORD review.

In addition to these activities designed to support NAAQS development and implementation, ORD's intramural and STAR grantees have developed and evaluated novel measurement methods to support other monitoring initiatives. Designed as a low-cost alternative to traditional integrated techniques, passive aerosol samplers have been developed to enable characterization of PM inorganic chemistry and morphology. Because the samplers operate in unattended mode and require no electrical power, they can be readily deployed at multiple sites to determine spatial variability of PM concentrations. Advances in real-time sampling and analysis techniques have enabled development of the Semi-continuous Elements in Aerosol System (SEAS) sampler for PM metals component determinations, the Ambient Ion Monitor (AIM) sampler for determination of PM anions and cations, and use of open-path optical techniques for determination of NO_2, SO_2, O_3, and hydrocarbon concentrations.

Impacts and Outcomes: The successful development and implementation of new reference and equivalent monitoring technologies provides vital tools to monitoring agencies for assessing and mitigating NAAQS pollutant concentrations. Development and evaluation of novel measurement technologies will support epidemiological and health studies, in addition to supporting development and implementation of new air quality regulations.

Future Directions: Air quality regulations for the criteria pollutants are continuously under review and ORD will continue to conduct laboratory and field studies of new monitoring methods to support proposed revisions of the NAAQS standards. While programmatic emphasis will be placed on making compliance measurements with minimal uncertainty to support compliance decisions, research initiatives will also focus on development and implementation of novel measurement techniques to support multiple monitoring objectives. In particular, development of near-real time monitors for speciation analysis of criteria pollutants will provide tools necessary to determine temporal variations of pollutant concentrations for supporting source apportionment studies, timely public advisories, and focused health studies.

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Poster # LTG 1-26

How have ambient measurements improved the understanding of secondary organic aerosol (SOA) formation?

Presenter: John H. Offenberg**,** US EPA Office of Research and Development

Summary: Fine particulate matter (PM2.5) is emitted directly into the atmosphere and formed through photochemical reactions of organic species in the atmosphere producing aerosol, which is known as secondary organic aerosol or SOA. SOA is an important part of PM2.5, especially during the periods of high photochemical activity. Current models used for air quality planning, such as the Community Multi-Scale Air Quality (CMAQ) model, significantly underpredict the concentrations of PM2.5. Improvements in our understanding of SOA formation to include all relevant precursors and major processes ensure that models represent the real atmosphere as well as possible.

Both field and laboratory studies improve our understanding of SOA formation processes and the sources of SOA precursors. Results from chamber experiments reveal previously unrecognized SOA precursors (isoprene, sesquiterpenes and benzene), the role of acidic aerosols and NOx concentration in SOA formation, and the importance of polymerization in producing SOA. Combining laboratory measurements of generated SOA mass fractions for a set of molecular tracer compounds for individual precursor hydrocarbons (isoprene, α -pinene, toluene, β -caryophyllnene) with tracer data measured in field samples from a variety of locations allows estimates of the SOA contributions for these precursors. Other field work using recent improvements to experimental protocols, determined the emissions of isoprene, sesquiterpenes, and monoterpenes from a variety of tree species, along with the temperature and light dependence of these emissions, which is essential to understanding how the emissions contribute to particulate matter concentrations under changing climatic conditions.

Impact and Outcomes: The results of this research provide insights regarding SOA formation in the atmosphere from a variety of anthropogenic and biogenic organic precursors. This work informs other research activities such as source apportionment and health studies and these results are being used to improve the SOA chemical mechanisms in air quality models, such as CMAQ. These models are used by EPA, states, and other air quality management organizations to develop and evaluate options for implementing air quality regulations. Incorporation of these advances in understanding will improve how well models represent particle concentrations in the atmosphere and increase the ability of planners to use these models to predict air quality and develop plans for reducing particulate matter concentrations.

Future Directions: Additional work will focus on field investigations of SOA formation under broadly dissimilar atmospheric conditions. Laboratory efforts will include 1) investigating the formation of inorganic and organic particulate nitrates, 2) examining the role of aerosol acidity on SOA formation from biogenic and anthropogenic precursor hydrocarbons, 3) further development of tracers for anthropogenic precursor hydrocarbons, 4) examining the role of fuels and fuel additive nano-particles on SOA formation, and 5) investigating the formation mechanism of SOA from aromatic hydrocarbons. Additionally, linked laboratory and field studies will improve our quantitative and mechanistic understanding of SOA production from oxidation of α- and β-pinene.

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Poster # LTG 1-27

How Can Measurements And Modeling Be Used To Improve The Understanding Of Mercury Fate And Transport?

Presenter: Jesse Bash, US EPA Office of Research and Development

Poster Summary: Environmental contamination from mercury has been recognized for decades as a growing problem to humans and wildlife. It is released from a variety of sources in chemically distinct different forms, exhibits a complicated chemistry, and proceeds via several different pathways to humans and wildlife. Atmospheric emissions and deposition processes are important drivers in mercury accumulation in soils and sediments, and are now recognized as the major route of contamination to most aquatic ecosystems. The most significant releases of mercury to the atmosphere in the U.S. are emissions from human activities, particularly the combustion of fossil fuels. While a plausible link has been made between mercury emissions and the presence of mercury in humans, a number of uncertainties related to its biogeochemical cycling remain.

EPA ORD is helping to reduce these uncertainties through an integrated program of measurements and modeling with a goal of improving information on the emission, transport, transformation and deposition of mercury in the environment to inform potential EPA policies. Measurement studies have focused on the development and evaluation of measurement methodologies, characterization of emissions from different source types, elucidation of atmospheric transformations, and source apportionment. Modeling research has been conducted through the development, evaluation, and application of the CMAQ model. The CMAQ model simulates mercury in three separate species: gaseous elemental mercury, reactive gaseous mercury, and particulate mercury and is updated as research findings on the chemistry of atmospheric mercury are published by ORD and other external organizations. Updated CMAQ simulations are then used to inform the research community of the scientific uncertainties to which the model is most sensitive, to guide future research endeavors.

Impacts and Outcomes: ORD's atmospheric mercury research program has contributed to the development of improved mercury measurement methods that have and will continue to be used to support emission characterization, atmospheric process, and source apportionment studies conducted by EPA and others. These methods have been applied in ORD's research program to enhance mercury emission inventories and air quality models and to conduct atmospheric and source apportionment studies that have provided insights into local, regional, and global mercury transport and fate. Furthermore, the CMAQ mercury model was used in the development of EPA's Clean Air Mercury Rule. It has also been applied in two mercury model intercomparison studies, one in Europe and one in North America. The second study demonstrated the importance of intercontinental transport and the need for accurate air concentration data at the regional model boundaries. Overall, ORD's atmospheric mercury research program is providing the Agency with state-of-science measurements and modeling tools to improve the understanding of local, regional, and global mercury fate and transport and to inform potential policies for the management of domestic mercury emissions.

Future Directions: Data from atmospheric mercury measurement studies will continue to be analyzed to inform model development. A new study in Cleveland, OH will provide new information on mercury deposition in an urban setting with multiple industrial sources. In addition, a new method for measuring mercury dry deposition will be evaluated in the field. The development and evaluation of the CMAQ model will continue including enhanced capabilities for bidirectional mercury exchange. In addition, the CMAQ mercury model will be tested and applied at the hemispheric scale. Finally, the measurements and modeling tools will be made available to inform potential EPA policy decisions.

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Poster # LTG 1-28

How do coarse particles vary regionally and within specific locales?

Presenter: Mike Hannigan, University of Colorado, Boulder, CO

Poster Summary: Time series studies have provided evidence of an independent effect of coarse particulate matter ($PM_{10-2.5}$) on morbidity and mortality. To date, the evidence suggests that the largest concern for $PM_{10-2.5}$ is an "acute exposure – morbidity effect" which is stronger than, or as strong as, that of $PM_{2.5}$ (Brunekreef et al., 2005). Since the atmospheric lifetime of $PM_{10-2.5}$ is inherently shorter than $PM_{2.5}$, we expect the temporal and spatial variability of the $PM_{10-2.5}$ to be greater than that of the $PM_{2.5}$. The inherent spatial and temporal variability of $PM_{10-2.5}$ and the acute exposure concern make $PM_{10-2.5}$ seem like an almost intractable problem. So, is it really that bad?

EPA-funded spatial and temporal characterization of $PM_{10-2.5}$ has occurred in Southern California, the Front Range of Colorado, Eastern North Carolina as part of the North Carolina Adult Asthma & Environment Study (NCAAES), Detroit as part of the Detroit Exposure and Aerosol Research Study (DEARS), Birmingham, AL, Gary, IN, Riverside, CA and Phoenix, AZ as part of the sampler equivalency studies.

The focus of $PM_{10-2.5}$ characterization during DEARS, NCAAES, and the current Birmingham study is the spatial variation in mass concentration. In Detroit, the $PM_{10-2.5}$ mass concentration was explored spatially during both summer and winter seasons. In the summer, all site pairs exhibited Pearson correlation coefficients greater than 0.33, in contrast to winter when some site pairs showed no correlation. The contrast is likely due to reduced mixing in the winter which will allow sites to be effectively impacted by local sources. There was also the suggestion that different sources may have different size distributions and as such if a source with a slightly smaller size distribution preferentially emits in the summer, than there will inherently be less spatial differences in the summer. Similar to DEARS but on a smaller spatial scale, investigators in NCAAES found that $PM_{10-2.5}$ mass concentrations measured at sites near sources were poorly correlated with sites not near sources, while the general trend was reasonable spatial agreement (14 of 18 sites with $r \geq 0.71$). In Birmingham, investigators are exploring spatial variability of $PM_{10-2.5}$ mass concentrations in two seasons. The use of dichotomous samplers allows for the potential to also explore composition variability as well although currently planned analyses are limited to mass concentration.

In Southern California, the Front Range of Colorado, and at three locations associated sampled during the equivalency study, both $PM_{10-2.5}$ mass concentration and composition were characterized. In Long Beach, CA, the spatial variability of composition was measured over a 10 week time period. Due to the mass needs of speciation, samples composites of 1-2 weeks were created. Spatial variability of $PM_{10-2.5}$ mass concentration was lower than that observed for any species. In other words, components exhibited more spatial variability than mass. In general, the composition of the $PM_{10-2.5}$ consisted of sea salt (31-55%), insoluble soil (11-47%), nitrate (14-23%), and organic matter (8-18%). Similarly, in Birmingham $PM_{10-2.5}$ sample collection time was long, 10 -16 days, as passive samplers were employed. Composition was measured with SEM and results showed soil, organic matter and possible some a Ca rich point source. In Denver daily $PM_{10-2.5}$ was characterized every 6^{th} day for an entire year. As seen in other sites, soil was the dominant contribution followed by organic matter. At the two of the three equivalency sampling sites, Riverside and Phoenix, a soil source is dominant followed by unidentified material. At Gary, local carbon sources are dominating the $PM_{10-2.5}$ material.

Impacts and Outcomes: These studies of $PM_{10-2.5}$ have started to illustrate the critical need to understand both temporal and spatial variations in relation to sources. Across communities and urban areas, when monitors are not placed near sources, there is reasonable spatial agreement in $PM_{10-2.5}$ concentrations. Composition of $PM_{10-2.5}$ is, not surprisingly, dictated by local sources. For example, coastal locations show a dominant sea salt contribution to $PM_{10-2.5}$. In addition, initial speciation efforts

illustrate a need to develop a new set of speciation protocols, as applying the $PM_{2.5}$ protocols to the $PM_{10-2.5}$ has made mass closure difficult. Both source and speciation thinking should inform the design of a $PM_{10-2.5}$ monitoring program.

Future Directions: Future research will focus on (1) understanding how urban (or smaller) scale variations in $PM_{10-2.5}$ mass concentrations are related to location of sources, (2) understanding temporal variation in $PM_{10-2.5}$ with hourly to daily resolution again in relation to source origin, and (3) understanding the regional difference in $PM_{10-2.5}$ composition that is dictated by differences in sources by region.

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Poster # LTG 1-29

How Have Atmospheric Chemical Kinetic Mechanisms Been Expanded for Multipollutant Atmospheric Modeling?

Presenter: Deborah Luecken, US EPA Office of Research and Development

Poster Summary: An accurate characterization of atmospheric chemistry is essential for predicting the response of air pollutants to emissions changes, characterizing spatial and temporal concentrations, and quantifying pollutant deposition. In the past, air quality modelers have largely focused on single pollutant issues (such as ozone), but it has since become clear that it is more appropriate to treat air pollution in an integrated, multi-phase, multi-pollutant manner. For example, both ozone and particulate matter respond non-linearly to reductions in NO_x, but are also affected by VOC/NO_x ratio. We also know that strategies to decrease ozone can have either beneficial or adverse consequences for hazardous air pollutants (Luecken and Cimorelli, 2008). The goal of our research is to develop, refine, and implement chemical mechanisms that can be used in the Community Multiscale Air Quality (CMAQ) model to accurately predict simultaneous concentrations of multiple pollutants that are of interest to the Agency, by:

- Representing interactions and feedbacks of important atmospheric air pollutants and their sensitivities to control strategies;
- Ensuring that CMAQ and other models used for regulatory and research purposes have scientifically-justifiable chemical representations, are appropriate for the application, and are consistent with our most up-to-date knowledge of atmospheric chemistry;
- Accounting for interactions between gas-, aqueous- and particle-phase chemistries, so that we can truly predict multi-media chemical effects of emissions changes; and
- Developing techniques and tools to efficiently expand current mechanisms to predict the chemistry of additional pollutants that we anticipate will become important in the future.

Our efforts are resulting in more complete and up-to-date descriptions of the important chemical pathways that influence concentrations of criteria pollutants. In single simulation, we currently predict simultaneous concentrations of ozone, particulate matter, nitrogen oxides and lead, as well as 43 Hazardous Air Pollutants (HAPs) including mercury.

Impacts and Outcomes: Our chemical mechanisms within CMAQ have been used by EPA during the past 5 years to predict potential benefits of a number of national programs such as the Clean Air Interstate Rule (CAIR) and the ozone NAAQS RIA (Regulatory Impact Assessment). The multipollutant chemical mechanism forms the basis of the EPA/OAQPS 2002 Multipollutant modeling platform, which is being used to examine potential co-benefits of regulatory actions such as the Renewable Fuels Standard Act (RFS2), which is examining how ethanol replacement of gasoline in automobiles might increase some HAPs, decrease others, and have different effects on $PM_{2.5}$ and ozone.

Future Directions: Because the chemistry impacts every component of air quality models, our future efforts in atmospheric chemistry mechanisms will continue to evolve and employ our expertise in gas, aqueous, and aerosol chemistry. Future efforts will involve reducing uncertainties that we know about in the current chemical mechanisms and improving the gas-aerosol-aqueous chemistry linkages. We anticipate that, given the breakthroughs in atmospheric chemistry in this field, we will also work to modify the mechanisms to include new information (such as new reactions) in order to keep our mechanisms at the state-of-the-science. We will extend the chemistry beyond “traditional” pollutants to address emerging issues such as biofuels, pesticides and chemicals that contribute to global warming.

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Poster # LTG 1-30

How Have PM Model Estimates Improved with Advances in Aerosol Process Representations?

Presenters: Prakash Bhave, US EPA Office of Research and Development
Michael Kleeman, University of California at Davis

Poster Summary: Accurate numerical predictions of particulate matter (PM) concentration, chemical composition, and size distribution are necessary for predicting the impacts of future air quality regulations and future climate on acute and chronic health effects, visibility degradation, acid and nutrient deposition, and climate change. The objective of this research is to improve predictions of PM in regional- and urban-scale models by advancing the scientific algorithms used to represent aerosol processes. To achieve this objective, research since 2005 has focused on three areas in which previous air quality models were deficient: secondary organic aerosol (SOA) formation, coarse PM, and ultrafine PM.

The most extensive model advancements were made in the treatment of SOA. Evaluation of previous model results against ambient measurements revealed persistent negative biases in the estimates of particulate organic carbon (OC) during summer and frequent positive biases during winter. The laboratory experiments of numerous investigators suggested that these biases could be driven by missing sources and/or incorrect thermal properties of SOA in the models. Subsequently, a number of modeling studies were undertaken to assess the importance of these laboratory-based findings. For example, the extent to which SOA can be produced by isoprene or sesquiterpenes and the influence of ambient NO_X concentrations on aromatic SOA formation were explored.

A number of model enhancements were made for treating coarse PM including a computationally-efficient scheme to treat the dynamic transfer of inorganic gases to and from coarse particles; extension of the ISORROPIA thermodynamic module to treat cations commonly found in crustal material; and adjustment of the size distribution of emitted sea-salt particles to account for ambient relative humidity. At the other end of the particle size spectrum, advances were made to simulate the ambient concentrations of ultrafine particles. These developments focused on capturing regional nucleation events and estimating the size distribution of emitted combustion particles.

Outside these three areas of emphasis, significant advancements were also made in tracking the source contributions to primary and secondary PM, simulating the trace metals within ambient PM, and improving the numerical stability of advection algorithms and thermodynamic processes.

Impacts and Outcomes: The revised SOA treatment has resulted in better predictions of aromatic SOA and the seasonal cycle of total OC, while capturing a new responsiveness of modeled SOA to changes in NO_X, SO_2, and NH_3 emissions. The enhanced treatment of coarse PM has resulted in better predictions of size-resolved nitrate in coastal areas which, in turn, improve the model estimates of nitrogen deposition in sensitive ecosystems. Many of the model enhancements described above have been incorporated into the Community Multiscale Air Quality (CMAQ) modeling system, which is used by Federal, State, and local authorities when making air quality management decisions and issuing health advisories (e.g., EPA Clean Air Interstate Rule, NOAA air quality forecasts). As a result, ORD's clients have increased confidence in the utility of CMAQ predictions for future regulatory applications (e.g., RFS-2 rulemaking). Finally, the impacts of research on coarse and ultrafine PM will be realized as the EPA considers new National Ambient Air Quality Standards (NAAQS) for those fractions of the PM size distribution.

Future Directions: In the near term, emphasis will shift toward simulating episodic events where the new 24-hour $PM_{2.5}$ NAAQS is exceeded. This may warrant investigations of the trace-elemental composition of $PM_{2.5}$, non-carbon organic material, and sampling artifacts associated with gravimetric $PM_{2.5}$ measurements. In addition, promulgation of new PM NAAQS and ongoing evaluations of the recently-revised air quality models will help prioritize future advancements in the aerosol process representations.

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Poster # LTG 1-31

How Do New Concepts of the Formation of Secondary Organic Aerosols Improve Our Modeling of Particulate Matter?

Presenter: Allen L. Robinson, Center for Atmospheric Particle Studies, Carnegie Mellon University

Poster Summary: Organic aerosol is a major component of fine-particle mass throughout the atmosphere and therefore organic aerosol concentrations must be addressed by regulations designed to reduce human exposure to fine particulate matter. Organic aerosol comprises primary organic aerosol (POA - particle mass directly emitted from sources such as motor vehicles and forest fires) and secondary organic aerosol (SOA - particle mass formed in the atmosphere from oxidation of gas-phase precursors). However, the relative importance of different sources of organic aerosol is uncertain -- even the POA-SOA split remains controversial. Recent field measurements indicate SOA dominance, even in heavily urbanized areas, that cannot be explained by the oxidation of known SOA precursors. The persistent discrepancies between measured OA concentrations and predictions of atmospheric chemistry models underscore the substantial uncertainty regarding the sources of organic aerosol.

Combining laboratory, field, and modeling results, we proposed a new conceptual model for primary organic aerosol that explicitly accounts for gas-particle partitioning of POA and allows gas-phase oxidation of all low volatility vapors in current SOA production mechanisms. This replaces the current, static representation of POA emissions with a far more dynamic picture in which low-volatility material evaporates, oxidizes, and recondenses over time. Dilution sampler and thermodenuder measurements demonstrate the semivolatile character of POA. Smog chamber experiments with diluted exhaust from real sources can only be explained with oxidation of low volatility primary emissions with high yields. These processes create a regional aerosol; implementing them in chemical transport models brings predictions into better agreement with observations.

To improve simulations of organic aerosol, we have developed a new unified framework of semi-volatile partitioning that permits models to efficiently treat both semi-volatile primary emissions and SOA production, and then to treat the chemical evolution (aging) of the aggregate distribution of semi-volatile material. The key feature of this treatment is a uniform basis set of saturation vapor pressures spanning the range of ambient organic saturation concentrations, from effectively nonvolatile material at 0.01 $\mu g\ m^{-3}$ to vapor-phase effluents at $10^6\ \mu g\ m^{-3}$. Chemical evolution is treated by a transformation matrix coupling the various basis vectors. The major advantages of the Volatility Basis Set (VBS) approach include: 1) it provides a single framework for describing gas-particle partitioning of both POA and SOA; 2) it allows for multiple generations of SOA chemistry without increasing the number of tracked SOA products; and 3) the required inputs – the volatility distribution of emissions and volatility operator – can be derived from experimental data.

Impacts and Outcomes: A better understanding of sources of organic aerosol allows development of more effective control strategies for fine particulate mass. The volatility basis set approach has been implemented into CMAQ and PMCAMx, two chemical transport models used by regulators to simulate regional air quality for State Implementation Plan (SIP) development for meeting air quality standards. The performance of these models has improved, especially with respect to predicting the level of oxygenated or secondary organic aerosol. The semivolatile character of POA means that we need to rethink how we measure emissions and represent them in models.

Future Directions: This work has raised many needs that must be addressed by future research in this area. First, we are developing a 2D-VBS with oxygen-to-carbon ratio (O:C) as the second dimension in addition to effective saturation concentration. Substantial research has begun to measure SOA yields from non-traditional low volatility precursors such as large n-alkanes and polycyclic aromatic hydrocarbons. We are working on understanding the SOA formation from multigenerational atmospheric chemistry.

Finally, research is needed to measure the volatility distribution of primary organic aerosol from important source classes.

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Poster # LTG 1-32

What is the Role of Atmospheric Mixed-Phase Chemistry in Multipollutant Modeling?

Presenter: Annmarie G. Carlton, US EPA Office of Research and Development

Poster Summary: Clouds cover ~60% of the Earth's surface and provide a means to transport constituents from the polluted boundary layer to the free troposphere, with substantial implications for long range pollution transport. Despite this predominance and importance, representation of cloud chemistry is minimal in most atmospheric models. Hundreds of explicit or semi-explicit kinetic expressions are used to describe gas-phase chemistry, while aqueous chemical mechanisms are usually limited to only a few (~5) sulfur oxidation reactions. These aqueous sulfur reactions contribute to acid rain and the formation of sulfate aerosol. Similarly, aqueous phase organic chemistry contributes to acid rain and organic aerosol also forms as a result of mixed phase chemistry.

Water-soluble products of the gas phase oxidation of alkenes and aromatics partition into atmospheric waters (e.g., cloud droplets and aerosol water) where they react further to form low volatility products. When this happens in clouds, these products remain, in part, in the particle phase after droplet evaporation, forming secondary organic aerosol (SOA). Zero-dimensional cloud chemistry and cloud microphysical modeling of in-cloud SOA formation have demonstrated kinetic feasibility. Laboratory experiments confirm the formation of low volatility compounds (e.g., oxalic acid and oligomers) during aqueous phase photooxidation of common atmospheric constituents. These low volatility compounds have been identified in aircraft measurements sampled from cloud-influenced air parcels and in collected rain water. Similar processes involving organic and inorganic (e.g., N_2O_5) compounds occur in aerosol water as well, adding to the atmospheric particulate matter burden.

Accurate prediction of SOA formed through mixed-phase pathways is hampered by the poor understanding of the complex aqueous phase organic chemistry, and by the dependence of product yields on cloud presence, cloud contact time, liquid water content and VOC/NO_x ratios. This poster will demonstrate the interactive process by which mixed phase chemical mechanisms can be evaluated and refined to generate atmospherically-relevant SOA yields and/or generate simplified chemistry for incorporation in chemical transport models. The poster will explore how improved representation of mixed-phase chemistry affects predicted atmospheric SOA mass concentrations and wet deposition amounts.

Impacts and Outcomes: SOA formation is typically modeled as reversible partitioning of semi-volatile VOC oxidation products. These formulations neglect mixed-phase chemical processes and tend to underpredict atmospheric concentrations of organic carbon (OC). Recently, addition of SOA production from cloud processing of glyoxal and methylglyoxal to the CMAQ chemical transport model has been shown to improve prediction of the vertical profiles of organic aerosol concentrations for a limited number of simulated days in the northeastern U.S. Similarly, an irreversible uptake process to simulate SOA formation through mixed phase processes in cloud droplets and other aerosols was added to the global model GEOS-Chem, and these simulations demonstrated enhanced representation of the variability in OC measurements. Improved predictions of OC, a substantial component of total aerosol mass, will ensure effective air quality management.

Future Directions: ORD will continue to work to improve the representation of mixed phase chemistry in the CMAQ model. A new generalized numerical solver for aqueous chemistry that allows for expansion of the current aqueous phase chemical mechanism has been developed and is currently being tested in CMAQ. Upon implementation of the new solver, a variety of mechanisms developed from box model simulations will be evaluated by comparing CMAQ-predicted atmospheric OC mass concentrations and speciated deposition amounts with observational data.

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Poster # LTG 1-33

How Do We Minimize Meteorological Model Uncertainties For Use In Air Quality Modeling?

Presenter: Jonathan Pleim, US EPA Office of Research and Development

Poster Summary: Air quality models require an accurate representation of air flow and dispersion, cloud properties, radiative fluxes, temperature and humidity fields, boundary layer evolution and mixing, and surface fluxes of both meteorological (heat, moisture, and momentum) and chemical species (dry deposition and evasion). Thus, meteorological models are critical components of air quality modeling systems. There is a need to reduce uncertainty in model results by development and improvement of physics process components and improvement of data assimilation/nudging strategies. Each of these research objectives for meteorological modeling has the overarching goal to lower error and uncertainty that translates directly to less uncertainty in air quality simulations. Our meteorology modeling research program involves several key projects that have led to improved meteorological fields. The first is the transition from an older mesoscale modeling system (MM5) to a more current model (Weather Research and Forecast model, WRF) that represents the current state-of-science. Part of this effort was to implement the land surface model (PX-LSM), surface layer (Pleim), and planetary boundary layer (Asymmetric Convective Model version 2; ACM2) schemes, which had been used in MM5, that are designed for use with retrospective air quality simulations. This work included enhancements of the PX-LSM that included a deep soil nudging algorithm and snow cover physics that dramatically improved temperature estimations in both the winter simulations and areas with less vegetation coverage. Another effort has been a re-examination of four dimensional data assimilation (FDDA) techniques including the development of an objective analysis program that serves to lower the error of analyses that are used to force the model towards observations. In addition, it is important that the surface processes and planetary boundary layer (PBL) modeling algorithms in the meteorology and air quality components of the system be as consistent as possible. For example, the PBL model, ACM2, is used in both WRF and CMAQ models.

Impacts and Outcome: Current results of the implementation of new physics and FDDA in WRF show that our configuration now exceeds the level of MM5 in terms of reduced uncertainty or error in surface variables like 2-m temperature, moisture and 10-m wind. A new evaluation method that utilizes both wind profiler and aircraft profile measurements provides a routine method to examine not only the uncertainty of simulated wind in the PBL but also the less examined temperature structure. The WRF model has low temperature error in the PBL and simulates the evolution of the wind structure, including features like nocturnal jets and the convective mixed layer with low error (< 2.0 m s^{-1}). CMAQ model results have been shown to benefit significantly, in terms of reduced ozone concentration error, when FDDA techniques are used in the meteorological simulation. Comparisons to data from the TexAQS II field experiment show good agreement with PBL heights derived from radar wind profilers and vertical profiles of both meteorological and chemical quantities measured by aircraft spirals. Increased fidelity of the meteorological data generated from the WRF model translates directly to increased confidence in the proper response of the CMAQ air quality model to changes in meteorology or source emissions.

Future Directions: A variety of data assimilation methods, including 3-D variational techniques (3Dvar), are being tested and evaluated to further reduce uncertainties in meteorology modeling. The 3Dvar system has the potential to improve simulated precipitation fields through the assimilation of satellite and radar data. The effects of meteorology model uncertainties on CMAQ model results are being investigated by evaluation of CMAQ simulations that are driven by different configurations of meteorology models. Improved models of the stable boundary layer (SBL), which is critical for the development of nocturnal inversion layers, low level jets, and steep concentration gradients near the surface, are being developed and tested.

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Poster # LTG 1-34

How Do Evaluation Techniques Establish the Credibility of Air Quality Model Estimates of Ambient Pollution Levels?

Presenter: Kenneth Schere, US EPA Office of Research and Development

Poster Summary: To advance the comprehensive model evaluation effort that is a critical component of the Community Multiscale Air Quality (CMAQ) model program, we present an evaluation framework describing the roles of operational, diagnostic, dynamic, and probabilistic evaluation approaches. Comparison of criteria pollutant predictions to observations (e.g., ozone, fine particulate matter [$PM_{2.5}$] mass and species) is a fundamental part of evaluation protocols, and it is critical to assess the role of various processes and model inputs to those predictions. These operational and diagnostic evaluation approaches can provide important insights to issues that can inform and improve the air quality model or the meteorological or emission inputs. Further, evaluating an air quality model's response to emission changes is central to how the model is used for air quality management at the local, state, and federal levels. The NO_x State Implementation Plan (NO_x SIP) Call offered a unique opportunity to conduct a "dynamic evaluation" of CMAQ's predicted O_3 change as a result of a large, abrupt NO_x emission reduction in 2004. Uncertainties in model inputs and processes are very difficult to characterize in deterministic models such as CMAQ, but having reasonable uncertainty estimates can provide additional help and guidance to the air quality management community. Probabilistic evaluation approaches are under development to characterize the impact of uncertainties in emissions, meteorology, and chemistry on air quality predictions using CMAQ and the CMAQ-Decoupled Direct Method (CMAQ-DDM). Credible uncertainty estimates can provide valuable information to air quality management decisions about the confidence in the predicted air quality changes and likelihood for reaching attainment of national air quality standards. Additionally, these uncertainty bounds can be useful to the research process by identifying model prediction errors falling outside the bounds of estimated uncertainty and that therefore warrant further investigation.

To assist in implementing the analyses suggested in operational model evaluations, an Atmospheric Model Evaluation Toolkit (AMET) has been developed and demonstrated. The AMET, built upon a relational database management system and statistical and graphical analysis programs, enables integrated model evaluation exercises across meteorological and air quality model results.

Impacts and Outcomes: The Model Evaluation Framework has been presented and discussed at several international conferences and in a review article currently undergoing peer-review, to provide leadership to the research community on conducting comprehensive model evaluation programs. The Framework has been further refined in air quality model evaluation workshops over the last two years involving many scientists from the international modeling community (AMS/U.S. EPA Workshop on the Evaluation of Regional-Scale Air Quality Modeling Systems, August 2007; U.S. EPA/Environment Canada/European Commission Workshop of the Air Quality Model Evaluation International Initiative, April 2009). The EPA Office of Air Quality Planning and Standards (OAQPS) has been using the AMET since 2006. AMET was publicly released in February 2008, and more than 200 users have downloaded AMET to date.

Future Directions: The Model Evaluation Framework continues to provide context for research planning in ORD/NERL's air quality modeling program. Research emphasis in coming years will focus on implementation techniques for demonstrating the newer components of model evaluation, namely probabilistic and dynamic model evaluation. Operational and diagnostic evaluation will be conducted on new applications of the CMAQ modeling system, including regional/continental scale applications, as well as emerging application areas on the fine (urban) and hemispheric scales.

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Poster # LTG 1-35

How can Air Quality Management Tools be used to Support Ecosystem Assessments?

Presenter: Robin Dennis, US EPA Office of Research and Development

Poster Summary: Atmospheric deposition of sulfur and nitrogen is a key contributor to ecosystem exposure and degradation, causing acidification of lakes and streams and eutrophication of coastal systems. Reductions in atmospheric deposition of sulfur and oxidized nitrogen due to human-health-driven regulations in the 1990 Clean Air Act (CAA) Amendments are expected to significantly benefit efforts to improve water quality. However, water quality managers are not taking advantage of information on anticipated deposition reductions in developing their management plans. Managers need to understand what to expect from atmospheric emissions and deposition. This understanding must come from an air quality model utilized as a laboratory; it cannot come from measurements. The goal is to bring air quality into ecosystem management through regional air quality modeling and to facilitate the air-ecosystem linkage.

Our approach is to collaborate with select, motivated air-water partners who are willing to work together to provide a test laboratory with the atmospheric model to explore, assess, and apply improved techniques to advance water quality management goals and test linkage approaches. We develop an understanding of the needs of the water quality managers through real-world experience/participation with model applications. Through identification of basic management questions, we define what research and tool developments for the air quality modeling system are needed to make the linkage functional and the air-ecosystem modeling applicable and useful. Results help provide answers to nearly universal questions uncovered in the course of the application studies: How much is depositing? Who and where is the deposition from? How much will deposition change due to CAA air quality regulations in the face of population and economic growth?

Impacts and Outcomes: Guidance and insights on several fronts has been developed. For example, modeling results have demonstrated that local solutions are not very effective and long-range transport dominates, so regional approaches are necessary to address ecosystem deposition of sulfur and nitrogen. In addition, the uncertainty in ammonia emissions/concentrations has been found to be very important. Modeling results have also indicated that emission reductions from CAA programs are expected to have significant impacts. This research is also having impacts in specific sensitive ecosystems. For example, air deposition reductions are now a vital component of Chesapeake Bay Program's restoration efforts. Critical air deposition information has also been provided to the Tampa Bay Estuary Program to address its Total Maximum Daily Load (TMDL) needs and assessment goals. In general, our efforts have opened the door for water quality managers to include air deposition and make their management plans more efficient and effective. The work has paved the way for using CMAQ in national NO_x-SO_x regulatory assessments to protect ecosystems, and for using CMAQ in U.S. critical loads analyses.

Future Directions:
- Work on nitrogen and precipitation budgets: reduce biases. Work on new science, e.g., NH3 bi-directional exchange.
- Extend tool capabilities: CMAQ DDM-3D (to other species) and Watershed Deposition Tool (WDT).
- Extend research to air-ecosystem linkages for national critical load mapping (terrestrial and aquatic).
- Investigate the impact of climate change on deposition.
- Extend applications to ORD's Ecosystem Services Research Program to contribute to ecosystem services assessments (e.g., biofuels).
- Investigate combining monitoring and model data into a single deposition field for assessments.

Relevant Publications

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April 2006: “Uncertainty Assessment of CMAQ Dry Deposition Predictions” Robin L. Dennis

April 2008: “Relative Responsibility Assessment of Sectors and States: Oxidized Nitrogen Deposition in 2020 (final numbers)” Robin L. Dennis

Letters of support (how support/information was used and viewed by the client):

From NPS to Robin Dennis (Shenandoah study)

From Chesapeake Bay on CMAQ usefulness to process

Holly Greening to Larry Reiter on usefulness of CMAQ results

Peer Reviewed Chapter in Process of being published as part of NARSTO Multipollutant Assessment; Chapter 8 by Christian Seigneur and Robin L. Dennis on Air Quality Modeling needs to support multipollutant accountability and critical load assessments.

Poster # LTG 1-36

How Can Air Quality Management Tools Be Used To Inform Climate Policy?

Presenter: Chris Nolte, U.S. Environmental Protection Agency, Office of Research and Development

Poster Summary: Since air quality is strongly influenced by weather, it is sensitive to climate change. Research conducted within the ORD intramural program and via STAR grants has examined ways in which climate change may impact air quality. These include changes in atmospheric circulation and stagnation, precipitation, chemical reaction rates, natural and human-caused emissions, and background concentrations. Global climate models (GCMs) have been used to simulate current climate conditions and the climatology at 2050 under different assumptions for how greenhouse gas (GHG) concentrations may evolve over the next few decades. These GCM simulations are then either used directly by global chemical transport models, or downscaled with regional climate models to drive regional air quality models focused on the continental U.S.

In addition, ORD is also developing a set of scenarios that represent plausible changes in U.S. regional GHG and criteria pollutant emissions through 2050. These scenarios account for factors such as population growth and migration, economic growth and transformation, land use change, climate change, energy resource availability and cost, technology change, and energy and environmental policies. Scenario assumptions are quantified using economic, land use, and energy system models. From the results, future-year emissions are projected, and resulting air quality is evaluated using emissions and air quality models.

Impacts and Outcomes: Results of sensitivity runs in which U.S. emissions are held constant indicate that climate change alone will lead to 1-10 ppb increases in surface ozone concentrations in polluted regions, with the largest effects in urban areas and during pollution episodes. This climate penalty implies that stronger emission controls will be needed to meet a given air quality standard than would be needed in the absence of climate change. Direct sensitivity calculations have shown that planned controls of emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) will continue to be effective strategies for air quality management. The effect of climate change on particulate matter (PM) concentrations, however, is more complicated and uncertain. Precipitation frequency and mixing depth are important drivers for changes to PM, but there is not yet consensus as to how these variables will change.

The development of detailed emission scenarios that more comprehensively account for changes in population, economics, land use, resources, technology, and policy, as described above, is ongoing. This work, however, has already led to a number of important outcomes. For example, ORD has developed the Integrated Climate Land Use Scenarios (ICLUS) model to project future population and land use. ORD also has developed energy technology databases for the MARKAL (MARKet ALlocation) energy system model. The MARKAL database has been released to the public, and ICLUS will soon be released. These tools are expected to be valuable resources to many groups that are developing and evaluating GHG mitigation and adaptation scenarios. ORD's MARKAL database currently is being used by a number of universities, governmental and nongovernmental organizations.

Future Directions: Work continues to reduce the uncertainty regarding the impact of climate change on ozone and particulate matter. A key question will be how best to reduce GHG emissions to mitigate climate change while maintaining and improving air quality. In fact, GHG mitigation provides the potential for large improvements in air quality, and ORD is working in partnership with OAR to provide the information and tools to aid decision makers in considering air quality co-benefits. ORD is continuing to develop the ICLUS model and MARKAL databases, and is using these tools to characterize emissions

scenarios. Further, ORD is also developing methodologies for using land use projections from ICLUS to spatially allocate future-year emissions.

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Poster # LTG 1-37

How Can Air Quality Management Tools Be Used To Improve Exposure Assessment?

Presenter: Vlad Isakov, U. S. Environmental Protection Agency, Office of Research and Development

Poster Summary: Historically, air quality modeling tools have been developed and applied to predict and analyze the impact of source emission reductions on ambient air concentrations, primarily in support of air quality standards implementation. However, because air quality models can provide more spatially and temporally resolved estimates of air pollutant concentrations, these tools are now being developed and applied to inform exposure assessments in support of air pollution health studies, which have traditionally relied on ambient air monitoring data. Furthermore, when air quality models are linked with exposure models that include data on human activities and indoor/outdoor pollutant relationships, an estimate of actual human exposures may be obtained that better accounts for human mobility and indoor exposure issues.

Detailed information on air quality is a key input to exposure models. In order to provide the best estimates of air concentrations, air quality modeling estimates should include local-scale features, long-range transport, and photochemical transformations. A new method has been developed to enhance air quality and exposure modeling tools so that they can address finer-scale air pollutant concentrations and exposures. The approach combines the results from two types of regional- and local-scale air quality models (CMAQ and AERMOD). The resulting hourly concentrations are used as inputs to exposure models to enhance estimates of urban air pollution exposures that vary temporally and spatially. Thus, the new method establishes a linkage between air quality and exposure modeling and will improve health assessments that include near-source impacts of multiple ambient air pollutants. We demonstrate how this linked air quality/exposure modeling approach may be used in future community-level environmental health studies by providing exposure estimates that reflect residences near large industrial facilities or major roadways. This research is an important component of an EPA feasibility study being conducted in New Haven, CT, that is examining the cumulative impact of various air pollution reduction activities (at local, state, and national levels) on changes in air quality concentrations, human exposures, and potential health outcomes in the community.

We have also developed a space-time statistical model that uses both monitoring data and air quality modeling results for predicting spatially/temporally resolved air quality concentrations. Using data fusion techniques provides more accurate air quality inputs to human exposure models such as SHEDS and APEX. The fusion models exploit the spatial information available in both air monitoring data and air quality model output and will lead to better prediction for the average personal exposure to a pollutant for demographic subgroups.

Impact and Outcomes: The application of air quality modeling tools has great potential to enhance the exposure assessments needed for health studies and risk assessments. Early findings have shown the value of more resolved air quality modeling results in characterizing community and individual level exposures. In addition, the combined use of air quality models and measurements is currently being evaluated as a means to measure the impact of local scale emission reduction activities on exposures.

Future Directions: Air quality modeling tools will be applied to inform exposure assessments for epidemiological studies investigating health effects of multiple air pollutants. In particular, air quality model predictions describing source-oriented PM component concentrations in multiple size cuts will provide new inputs to examine the effects of acute and chronic PM exposure on mortality and morbidity.

Relevant Publications

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Poster # LTG 1-38

How ORD Air Research Helps Inform the Multi-pollutant Review of a Secondary National Ambient Air Quality Standard (NAAQS) for Oxides of Nitrogen and Sulfur (NOx and SOx)

Presenter: Anne W. Rea, US EPA Office of Air Quality Planning and Standards, Office of Air and Radiation

Poster Summary: EPA is currently conducting a multi-pollutant review of the existing secondary, or welfare-based, national ambient air quality standards (NAAQS) for oxides of nitrogen (NOx) and sulfur (SOx) together. Section 109(b)(2) of the Clean Air Act defines the purpose of secondary NAAQS to be to protect the public welfare from any known or anticipated adverse effects associated with the presence of the criteria air pollutants in ambient air. Because NOx and SOx pollutants are intricately linked from their emissions stages through their atmospheric transformation and concentrations to their deposition and ecological effects, the science, risks, and policies relevant to protecting the public welfare associated with these two pollutants are all best assessed for NOx and SOx together. In addition, data on the emissions, atmospheric science, and ecological effects of ammonia and ammonium are assessed since these interact tightly and pervasively with oxidized forms of nitrogen. Acidification due to both nitrogen and sulfur deposition and nutrient enrichment from nitrogen deposition are the targeted aquatic and terrestrial ecological effects considered in this review. Products from ORD's Air Research Program including work on the interactive nature of the many reduced and oxidized forms of nitrogen have been uniquely beneficial for addressing key questions in this NAAQS review, the first to consider these multiple pollutants together for the secondary standard.

Question 1: What is the relationship between the ambient air concentrations of these pollutants and their ecological effects?
The Community Multi-scale Air Quality modeling system (CMAQ) from ORD's Air Research Program is the primary tool used in this NAAQS review to connect pollution emissions and atmospheric concentrations with deposition to the Earth's surface where they can have biological effects. Pollutant emissions, chemistry, and CMAQ itself were assessed as part of the NOx and SOx Integrated Science Assessment (ISA) produced by ORD's National Center for Environmental Assessment (NCEA). CMAQ predictions for dry deposition are then combined with estimates of measured wet deposition at the National Atmospheric Deposition Program (NADP) sites to produce deposition fields for nine case study locations. This work has allowed OAQPS to better characterize and understand how nitrogen and sulfur species emitted in one region of the U.S. interact and travel to be deposited in another, and to conduct regional sensitivity analyses with the model to test that understanding.

Question 2: What are the relative contributions to observed ecological effects from total oxidized sulfur, from total nitrogen, from all oxidized forms of nitrogen, and from reduced forms of nitrogen?
Additional work with CMAQ has allowed OAQPS to estimate the relative responsibilities of the various components of total nitrogen and total sulfur deposition for ecological effects in developing concentration-deposition trade-off curves to evaluate a potential new multi-pollutant ambient secondary standard. This work has required incorporation of some of ORD's most recent work on ammonia emissions and atmospheric transformation in order to correctly attribute its contribution to the combined effects observed in affected ecosystems.

Question 3: How can we adequately characterize an adverse effect on public welfare in the framework of a secondary NAAQS?
The interface between ORD's Air Research Program and Ecosystem Services Research Program (ESRP) has benefited this multi-pollutant NAAQS review. The deposition estimates created using CMAQ have been used as the input data for several ecological models. The ESRP has played an important role in

conceptualizing the role of ecosystem services and their ability to characterize an adverse effect to public welfare. The ESRP is specifically developing a nitrogen research strategy which will focus on the ecosystem services associated with reactive nitrogen.

Impacts and Outcomes: Research from the ORD Air Program has supported OAR's progress and development on the current secondary NOx and SOx NAAQS review. While data are limited, the CMAQ+NADP hybrid deposition estimates have been used as the input data for the ecological effects modeling required to characterize effects in a way that will support decisions about a potential national ambient air standard. In addition, these CMAQ+NADP hybrid deposition estimates have been used to evaluate the relative contributions of oxidized and reduced forms of nitrogen. OAR has also relied on the Long-Term Monitoring (LTM) and Temporally Integrated Monitoring of Ecosystems (TIME) programs designed and begun by ORD to look at long term trends and surface water concentrations of SO_4^{2-}, NO_3^-, and the acid neutralizing capacity (ANC) of lakes and streams. ORD research continues to contribute to the scientific foundation being used to set an ecologically meaningful NOx and SOx Secondary NAAQS. This standard has been supported by ORD's Air Research Program with CMAQ modeling and multi-pollutant air chemistry research and by the nitrogen-based research from ORD's Ecological Services Research Program in close connection with the Air Research results. In addition, ORD NCEA has assessed and synthesized the science – much of it produced by the Air Research Program, and determined the level of effects causality for groups of ecological effects in their ISA.

Future Directions: CMAQ will continue to be applied to assess deposition of air pollutants associated with the NAAQS and Hazardous Air Pollutant (HAP) programs. ORD's Ecological Services Research Program (ESRP) is currently drafting an integrated nitrogen planning document to guide implementation of their research on reactive nitrogen effects and this effort is being supported by scientists from the ORD Air Program and partners from OAQPS. The work completed for this secondary NAQQS Review that integrates ORD modeling and measurements on multiple atmospheric pollutants with OAR policy needs has been very instructive to the ESRP planners as they defined policy-relevant questions about the interactions and effects of reduced and oxidized nitrogen. This creates a substantial new opportunity to integrate research from ORD's Air Research Program with that of the ESRP to help ensure that the most policy-relevant research can contribute to policy reviews of ambient standards.

Poster # LTG 1-39

Example Uses of the Community Multiscale Air Quality (CMAQ) Model to Support the Development of OAR Regulations and Air Quality Management

Presenter: Norm Possiel, US EPA Office of Air Quality Planning and Standards

Poster Summary: Effective air quality management at the national and local level is strongly dependent on the ability to reliably project the potential impact of specific air pollution control measures. In recent years, numerous OAR regulations and policies have been supported by ORD's Community Multiscale Air Quality (CMAQ) model. CMAQ simulations have been used to estimate the air quality improvement expected from national control programs to help areas attain National Ambient Air Quality Standards for ozone and fine particles, as well as to guide the design of control strategies that would most effectively improve air quality and reduce exposure. This poster discusses two on-going CMAQ applications at different scales (i.e., national and local) that will be used to inform air quality decisions in the near future.

EPA is required by Executive Order 12866 to conduct national Regulatory Impact Analyses (RIAs) to quantify the costs and benefits of major Federal programs designed to protect public health and welfare. In support of the RIAs, CMAQ is used to project air quality for future baseline and control case emissions scenarios associated with specific rules.
OAR has recently completed a national-scale air quality modeling analysis with CMAQ to analyze the impacts of the expect impacts of emissions reductions associated with the Locomotive and Marine Engine Rule. The results of this analysis are presented in the poster.

In response to the NRC Report, OAQPS undertook the Detroit Multi-pollutant Pilot Project to demonstrate an approach for considering multiple pollutants in an integrated manner for air quality planning at a local scale. Based largely on EPA's 2002-based multi-pollutant CMAQ platform, the study covered the Detroit metropolitan area and compared a comprehensive Multi-Pollutant, Risk-Based (MPRB) emissions reduction strategy to a traditional single pollutant approach to the design of control strategies. CMAQ was used to produce air quality concentrations of ozone, fine particles, and several key air toxic species across the Detroit urban area for two alternative control strategies. The results indicate that the Multi-Pollutant, Risk Based emissions reduction strategy achieved greater regional and local air quality improvement, compared to the single-pollutant approach.

Impacts and Outcomes: Results from our national rule modeling are used to project the air quality improvement in ozone and fine particle nonattainment areas and as input to downstream benefit and human health models to quantify the benefits associated with the rule. The Detroit Multi-pollutant Pilot Project illustrates the benefits of a comprehensive Multi-Pollutant, Risk-Based emissions reduction strategy relative to a traditional single pollutant approach, and demonstrates an approach for considering multiple pollutants in an integrated manner for air quality planning. This pilot project pointed to the importance of having a multi-pollutant version of CMAQ for understanding the impact of control strategies on multiple pollutants and human health outcomes.

Future Directions: OAR plans to expand modeling capabilities to address global scale air quality issues (e.g., international transport and climate), as well as fine-scale and near-source population exposure to pollutant concentrations. We will use CMAQ to assess the co-benefits air quality changes from the use of alternative fuels and other potential climate-related pollutants. The results from the urban scale Detroit Multi-Pollutant Pilot Project will be used to inform state, tribal, and local agencies interested in developing a comprehensive Air Quality Management Plan and to support plans for a National Air Pollutant Assessment (NAPA) which will characterize concentrations of ozone, particulates, various toxic species, and other pollutants.

Relevant Publications

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Poster # LTG 1-40

ORD Mobile Source Emissions Research Provides Data to Improve EPA Models and Regulatory Decision-Making

Presenter: Rich Cook, US EPA Office of Transportation and Air Quality, Office of Air and Radiation

Poster Summary: ORD's mobile source emissions research contributes to the development of key EPA models and the development of EPA's mobile source regulatory programs. Recent emissions research programs include measurements from on-road motor vehicles and non-road engines such as lawn and garden equipment.

For motor vehicle emissions testing, OAR and ORD partnered with state and local environmental agencies, the Department of Energy's National Renewable Energy Lab, the Federal Highway Administration, and the Coordinating Research Council) to conduct the Kansas City Light-Duty Vehicle Emissions Study. The study characterized the distribution of PM emissions from the on-road, light-duty motor vehicle fleet in the U.S., and also speciated gaseous and PM emissions from the in-use fleet. In addition, demographic data were collected on all vehicle owners. Exhaust emissions were measured during summer and winter from over 500 in-use consumer cars in the Kansas City metropolitan area. Results demonstrated the impacts of multiple factors on vehicle emissions, most notably vehicle technology/model year, mileage, and ambient temperatures. PM emission rates ranged over three orders of magnitude across the tested fleet; while select vehicles tested during the summer and winter yielded increased emission rates of an order of magnitude at colder ambient temperatures. In addition, the large amount of demographic data collected in this program will enable EPA to better understand the relationship between socioeconomic factors and motor vehicle pollution. Also, speciation data are being used in conjunction with improved chemical mechanisms in CMAQ to model formation of soluble organic aerosols, and will inform identification of pollutants with potential health concerns for toxicity evaluation.

Results from the Kansas City Study also promoted concerns that advanced technology vehicles meeting the most recent emission standards were emitting much higher levels of hydrocarbon emissions that estimated by the MOBILE6 emission factor model during starts at cold temperatures. Thus, substantial reductions in air toxic emissions could potentially be achieved through cost-effective control of start emissions. Recently collected data from ORD motor vehicle emissions testing elucidated the relationship between emissions of total VOC and gaseous air toxics at various temperatures. Additional testing was conducted on a flexible-fueled vehicle to better understand the impact on emissions of different gasoline-ethanol blends (E10, E20, E85) at different ambient temperatures for different soak times. Test program results showed that emissions of benzene and 1,3-butadiene increased proportionally with hydrocarbon emissions, with a strong correlation, whereas the correlation was not as strong with aldehydes. Furthermore, testing on the flexible-fueled vehicle indicated that the total hydrocarbon emissions did not increase linearly between 0 and 75 degrees Fahrenheit, and that soak time had a significant impact.

In addition to on-road motor vehicle emission testing, ORD conducts research on the emissions and exposures from the use of non-road engines. For example, ORD measured emissions from in-use and new technology lawn and garden equipment, as well as exposures to users of this equipment from exhaust emissions. Results indicated that emissions led to exposures of public health concern. These data helped form the justification for additional emission control standards for this equipment category.

Impacts and Outcomes: Results from the motor vehicle emissions studies help form the foundation for EPA's MOVES highway vehicle emissions model, providing estimates of rate and composition of particulate matter and gaseous emissions from motor vehicles. In addition, speciated emissions data

allow estimates of toxicity as well as precursor emissions for photochemical modeling. Moreover, real-time emissions data collected in the program has enabled EPA to characterize emissions under different operating modes to determine impacts from changing activities such as idling, acceleration, and high speed operations that illuminate potential emission impacts in near-road environments. These advances in emissions characterization are informing decision-making in several ongoing regulatory activities. Improved understanding of ambient temperature effects on emissions will lead to better emission inventories and control strategies. The non-road emissions and exposure study also provided information on the costs and benefits of implementing emission control strategies.

Future Directions: Additional motor vehicle emissions data will be needed to improve estimates of PM, VOC, and toxic emissions in MOVES to represent impacts from the U.S. fleet under multiple vehicle, fuel, and environmental factors. Data from these studies form a baseline for comparison to emissions data being collected on advanced technology vehicles and alternative fuels. Understanding the relationship between motor vehicle and non-road engine emissions and impacts on public health and the environment require a continuing understanding of the relationship between vehicle type/technology, vehicle activity, fuel composition, environmental conditions, and resulting emissions. This understanding is imperative for EPA to continue to develop efficient and cost-effective programs mitigating impacts from mobile source emissions. In addition, more data are needed on greenhouse gas emissions from motor vehicles to better inform climate change mitigation strategies.

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